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THE EFFECT OF DIFFUSIVITY ON GAS–LIQUID MASS TRANSFER IN STIRRED VESSELS. EXPERIMENTS AT ATMOSPHERIC AND ELEVATED PRESSURES

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Abstract—Mass transfer has been studied in gas–liquid stirred vessels with horizontal interfaces which appeared to the eye to be completely smooth. Special attention has been paid to the influence of the coefficient of molecular diffusion. The results are compared with those published before. The simplifying assumptions of identical hydrodynamical conditions at the same stirrer speed in one particular geometry, which have been made in some previous investigations, is shown to be wrong and may lead to incorrect conclusions on the influence of the diffusion coefficient. For the gas phase the mass transfer can be described by the penetration theory (Higbie, R., 1935, *Trans. Am. Inst. Chem. Engrs* **35**, 36–60) or surface renewal model (Danckwerts, P. V., 1951, *Ind. Engng Chem.* **43**, 1460–1467). With the use of a dimensionless equation, *Sh*, *Re* and *Sc* numbers, all data, even experiments carried out at elevated pressures, could be well correlated. For the liquid phase the results indicate that the mass transfer cannot be described by a simple model. The King model (King, C. J., 1976, *Ind. Engng Chem. Fundam.* **5**, 1–8), a combination of molecular and eddy diffusivity, is able to explain qualitatively the observed phenomena and the literature data.

1. INTRODUCTION

Many industrial chemical processes involve mass transfer between gas and liquid phases. In the past, a number of different approaches to the description of mass transfer in gas–liquid systems have been proposed, of which the film model (Whitman, 1923) and the penetration model (von Wroblewski, 1877; Higbie, 1935; Danckwerts, 1951) are most often applied. Although the various models are based on different descriptions of the mass-transfer process, the important difference between the various models can be found in the dependence of the mass-transfer coefficient on the diffusion coefficient. For example, the film model predicts that the mass-transfer coefficient, k_m, is proportional to the diffusion coefficient, D, and the penetration model predicts that it is proportional to $\sqrt{D}$. Several more complex models predict different dependences on the power of D.

Although probably none of the various models will give an accurate description of the mass transfer under all process conditions, it can be important for specific cases to know which model will cover the influence of diffusivity over a wide range. For example, for the absorption of a gas into a liquid, in which a reaction instantaneous with respect to mass transfer takes place, the nature of the model applied has a great effect on the predicted overall mass-transfer rate if the diffusivity of the liquid-phase reactant differs considerably from that of the dissolving gas. Also for the gas-phase controlled mass transfer of a system which is operated at various pressures the model applied determines the outcome of the mass-transfer calculations.

At our department the selective removal of H_2S from sour gases by reactive liquids is one of the subjects of research effort. For this type of process it is important to know which model covers the description of mass transfer best. The reaction between H_2S and the alkaline agent in the scrubbing liquid, often an alkanolamine (Kohl and Riesenfeld, 1981), is extremely fast and can be regarded as a reaction instantaneous with respect to mass transfer. Moreover, the ratio of the diffusion coefficients of the two reacting components differs considerably from unity. For design purposes, but also for the accurate interpretation of laboratory experiments, a good description of the mass-transfer rate in the liquid phase is required. As in many studies stirred vessels are used in our department in order to obtain accurate and reliable data on gas- and liquid-phase transport properties.

As pointed out above, for gas-phase mass transfer a good model is also needed, since on the one hand, most experimental work is carried out at atmospheric conditions while the industrial processes are carried out at elevated pressures (up to 10 MPa) and on the other hand, the influence of pressure on the mass-transfer behaviour of gas–liquid contactors has hardly been studied. Only a few publications on the influence of elevated pressures on gas–liquid mass transfer are known (Shridhar and Potter, 1980; Teramoto *et al.*, 1974; Vafopoulos *et al.*, 1975; Yoshida and Arakawa, 1968). However, generally it was not possible to predict the gas-phase controlled mass-transfer rates with the resultant data because the experiments were limited to measurement of k_1S or k_1 in stirred vessels and, moreover, some of the results were contradictory.

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Both for the liquid-phase mass-transfer coefficient, k_l, and the gas-phase mass-transfer coefficient, k_g, information on the influence of diffusivity is available in the literature. However, the results are not well in line and sometimes confusing. Therefore the available experimental data are critically reviewed and new data are presented on the influence of diffusivity on the mass-transfer coefficients both in the gas and liquid phase. The experiments were carried out in a stirred vessel with a horizontal, smooth gas–liquid interface. Therefore the interfacial contact area is well defined and known, and contrary to other contactors such as bubble columns and packed bed reactors the mass-transfer coefficients can be measured directly in a stirred vessel. Similarly in the near future different contactors will be studied.

2. REVIEW

2.1. *Mass-transfer models*

All models developed in the past for the description of the rates of mass transfer have in common that one of the transport mechanisms is molecular diffusivity and is described by Fick's law. The more complex models often include more transport mechanisms, e.g. eddy diffusivity. Each group of models has a different dependence of the mass-transfer coefficient on the diffusivity. Another essential distinction between the models is the mechanism of replacement of the stagnant fluid elements at the interface. Characteristic for each group of the models is the number of parameters which are necessary for the calculation of the mass-transfer rate. The models can be divided into three categories.

The first group consists of film models in which mass transfer takes place by stationary molecular diffusion through a stagnant film only as was proposed first by Whitman (1923). It is assumed that a stagnant film of thickness δ exists at the interface, while the bulk of the liquid is well mixed. For these models the mass-transfer coefficient is proportional to the coefficient of molecular diffusion D if the film thickness can be considered to be independent of the diffusion coefficient.

The second group of models is based on surface renewal. A stagnant fluid element is exposed at the interface for a finite period of time and the mass transfer can be regarded as non-stationary molecular diffusion into an infinitely deep stagnant medium. The replacement of the elements at the interface can be brought about by turbulent motion of the body of the fluid or by creation of a new interface when the fluid runs down over the surface of a packing or discontinuity. The characteristic parameter is the exposure time of the fluid elements at the interface. Higbie (1935) proposed that all elements have an identical residence time, θ, at the interface, while Danckwerts (1951) introduced a probability of replacement, s. The surface renewal type of models predicts k_m to be proportional to $\sqrt{D}$ if the exposure time is independent of D.

The third category consists of multi-parameter models. Two-parameter models which are a combination of the surface renewal and stagnant film approach have been developed by Dobbins (1956) and Toor and Marchello (1958). They assumed finite dimensions of the fluid elements replaced and their approaches can be regarded as a film-renewal model with two parameters, δ and θ. The dependence of the mass-transfer coefficient ranges from proportional to $\sqrt{D}$ up to proportional to D. Another type of multi-parameter model (King, 1966; Harriott, 1962; Kishinevskii and Pamfilev, 1949; Kishinevskii, 1955; Kishinevskii and Serebrianskii, 1956) describes mass transfer as a combination of molecular diffusion and interfacial turbulence which is regarded as eddy diffusion. The eddy diffusivity is assumed to be proportional to a certain power of the distance, perpendicular to the gas–liquid interface. The resulting influence of the molecular diffusivity on the mass-transfer coefficient is proportional to D^r, where $0.5 \leqslant r \leqslant 1$.

In all of the mass transfer models the parameters δ, θ and s are affected by the hydrodynamic conditions and physical properties of the system. In studying the influence of diffusivity on mass transfer, it is very important to take the hydrodynamics of the experimental systems used properly into account. If not, part of the influence of diffusivity on the mass-transfer coefficient can be disguised by hydrodynamical effects.

2.2. *Review of the literature*

Comparison of the literature data indicates that there are considerable discrepancies between the various studies with respect to the dependence of the mass-transfer coefficient on the diffusivity. Usually the results are presented as

$$k_m \propto D^r \tag{1}$$

or for reactors where the interfacial area is not known or measured separately:

$$k_m S \propto D^r. \tag{2}$$

By presenting the results in this manner it is implicitly suggested that the influence of the hydrodynamic properties of the system used are known or at least kept identical. However, in some of the studies this suggestion is not correct and therefore part of the scatter in the results can be explained by changes of the systems other than the variation in diffusivity of the component to be transported alone.

In Table 1 the literature sources considering the investigations in stirred vessels are summarized. Experimental data on the gas-side mass-transfer coefficient in stirred vessels has been published by two authors only. Tamir and Merchuk (1978, 1979) studied the desorption of pure liquids into various inert carrier gases. They state that identical hydrodynamic conditions were maintained in all experiments by keeping the stirrer speed, the gas flow rate and the height of the stirrer above the liquid surface constant. The validity

Table 1. Literature data on mass transfer in stirred vessels

Reference	System	Phase†	Exponent r
Tamir and Merchunk (1978, 1979)	Evaporation of pure liquids in various carrier gases	G	0.684 0.632
Yadav and Sharma (1979)	Absroption of solutes into aqueous solutions in which an instantaneous reaction occurs	G	0.50
Davies *et al.* (1964)	Absorption of H_2, He, O_2 and CO_2 into water	L	0.62
	Ar and He into water	L	$0.5 < r < 0.6$
	Ar and He into water or aqueous electrolyte solutions		0.50–0.67
Hikita and Ishikawa (1969)	Absorption of C_2H_4, CO_2, O_2 and H_2 in H_2O at 25°C and absorption of CO_2 in various aqueous solutions	L	0.67
Kozinsky and King (1966)	Absorption of He, H_2, O_2, Ar and CO_2 in H_2O at 28°C	L	0.50–0.67
Linek *et al.* (1970)	Absorption of He, N_2, O_2 and Ar in H_2O and aqueous electrolyte solutions	L	0.50–0.67
Machon and Linek (1974)	Absorption of He and O_2 in water and aqueous electrolyte solutions	L	0.67
Tavares da Silva and Danckwerts (1968)	Absorption of H_2S into aqueous alkanolamine solutions	L	0.50
Kafarov *et al.* (1978)	Physical absorption of H_2, N_2, O_2 into water	L	$0 < r < 1$

†G, gas phase; L, liquid phase.

of this statement is doubtful as the *Re* number [defined as eq. (7)] varied drastically (a factor of 15) in their experiments due to the different properties of the fluids used (see their Table 1) and therefore the hydrodynamic conditions may have changed considerably. Also questionable is the use of the viscosity and density of the carrier gas for the calculation of the dimensionless numbers. Small amounts of solute may change the physical properties of the gas mixture, and hence the hydrodynamic conditions, drastically. For instance, in their experiments of the desorption of CCl_4 in He as a carrier gas minute amounts of CCl_4 (0.03 mole fraction was a minimum value according to the authors) in He increased the gas-phase density by a factor of 2 and, therefore, their dimensionless numbers were not calculated correctly. The conclusion that $r = 0.684$ therefore does not seem well founded on their experimental results. The same remarks are applicable to the work of Yadav and Sharma (1979) and indicate that their conclusion ($r = 0.50$) is also questionable.

With respect to the liquid-side mass-transfer coefficients, the literature sources can be divided into three groups. Firstly, the investigations where the variation of diffusivity is realized by using different solutes; secondly, by a variation of the liquid properties; and thirdly, by a combination of the first two methods.

Davies *et al.* (1964), Kozinsky and King (1966) and Linek *et al.* (1970) studied the absorption of several solutes into water at a constant stirrer speed. The hydrodynamic conditions for these experiments can be considered constant at a given agitation rate. There is excellent agreement between these studies on the dependence of mass transfer on diffusivity with $r = 0.50$. Introduction of surfactants (Davies *et al.*, 1964) or inorganic electrolytes (Linek *et al.*, 1970) changes the value of r from 0.50 to 0.67.

Tavares da Silva and Danckwerts (1968) studied the absorption of H_2S into very dilute aqueous solutions of different amines, thereby keeping the hydrodynamic conditions of the systems constant resulting in $r = 0.50$. It should be noted that with this technique the authors measured the influence of diffusivity on the mass-transfer rate for the transport of a component in the liquid bulk to the gas–liquid interface. Hikita and Ishikawa (1969) and Machon and Linek (1974) varied both the solute and the liquid composition. The data were correlated by dimensionless numbers:

$$\text{Hikita and Ishikawa:} \quad Sh = 0.322\, Re^{0.7}\, Sc^{0.33} \tag{3}$$

$$\text{Machon and Linek:} \quad Sh = 1.90 \times 10^3\, Re^{1.01}\, We^{-2.19}\, Fr^{-1.98}\, Sc^{0.36}. \tag{4}$$

By presenting the results in this way a distinction of the influence of the hydrodynamic conditions and diffusivity on the mass-transfer coefficient is possible and from eqs (3) and (4) it follows that $r = 0.67$. Hikita and Ishikawa (1969) also presented results on the absorption of CO_2, C_2H_4, O_2 and H_2 into water at a constant stirrer speed. For these experiments the value of $r = 0.67$ was equally found.

Overall it can be concluded that the influence of diffusivity on liquid-phase mass transfer for the stirred vessel was found to vary from 0.50 to 0.67. For the gas phase it is not possible to give a general conclusion. It is very important that the hydrodynamics of the exper-

imental systems used are thoroughly taken into account in order to allow a separation between the influence of diffusivity and hydrodynamics on mass transfer. Additional experiments, as will be presented in this paper, seemed appropriate to clarify further the influence of the diffusion coefficient on the mass-transfer rate in stirred vessels with a smooth interface, in particular for gas-phase controlled mass transfer at higher pressures for which data are extremely scarce.

3. EXPERIMENTAL

3.1. *General remarks*

As stated before, the influence of the hydrodynamics on the mass-transfer rate should be known in order to be able to study the influence of diffusivity properly. Therefore it is important to use an experimental set-up of which the hydrodynamics can be easily studied and controlled. For this reason a stirred vessel, operated under conditions with a smooth gas–liquid interface, was chosen in our work. Apart from the gas pressure and physical properties of the fluids used, the only experimental parameters were the stirrer speed, geometric factors (like the position of the stirrer) and the residence times of the phases. The well-known type of dimensionless equations for mass transfer were used (Hikita and Ishikawa, 1969; Gilliland and Sherwood, 1934) to correlate the data:

$$Sh = c1 + c2Re^{c3}Sc^{c4}. \quad (5)$$

Here, the dimensionless numbers are defined as:

$$Sh = k_m d_s/D \quad (6)$$

$$Re = (d_s)^2 N\rho/\eta \quad (7)$$

$$Sc = \eta/\rho D. \quad (8)$$

The *Re* number is based on the stirrer speed which is valid only if the liquid and gas throughput in the stirred vessel is relatively low. This has to be investigated separately.

At relatively high *Sh* numbers, the exponent of the *Re* number in eq. (5) can be easily determined experimentally by the variation of the stirrer speed, as the value of the constant $c1$ can be neglected. The exponent of the *Sc* number can be evaluated by using different solutes at a constant *Re* number (indicating identical hydrodynamic conditions). The validity of eq. (5) can be further checked for the gas-phase mass transfer by means of variation of the system pressure, because for a pressure change over a not too wide range, the *Sc* number remains constant. Therefore the experiments for one particular system carried out at different pressures can be correlated by only one single equation.

3.2. *Measurement of the gas-phase mass-transfer coefficient*

The measurement of k_g was carried out by absorption of a dilute single gas component from an inert gas phase into a liquid where an instantaneous reaction occurred between the absorbed component and a liquid-phase reactant.

By correctly choosing the experimental conditions (see Danckwerts, 1970), completely gas-phase controlled mass transfer could be achieved. The variation in diffusivity was obtained in three ways: firstly, by changing the total pressure of the system; secondly, by using different inert carrier gases; and thirdly, by altering the reactive component in the gas phase.

The experimental set-up is shown in Fig. 1. The experiments were carried out in an 88 mm i.d., 1.22 l stirred glass vessel. Gas-phase agitation was provided either by two turbine impellers on one shaft (4 blades, 52.5 mm diameter, 15 mm height, 15 mm spacing, 8.3 mm diameter shaft, 85 mm shaft length) or by two propeller-type impellers (4 blades, 50 mm diameter, 5 mm height, 43 mm spacing). The liquid level was maintained at 35 mm below the lower gas-phase stirrer. In order to achieve a good distribution, the gas was introduced and distributed by means of a 68 mm Teflon (5 mm i.d.) tube with 25 holes (1 mm diameter). The interfacial area was 6.1×10^{-3} m^2. The gas-phase residence time ranged from about 24 s to up to 72 s. The gas-phase stirrer speed was varied between 0.45 and 26.7 rps, i.e. a range in the *Re* number from 200 to 26,000.

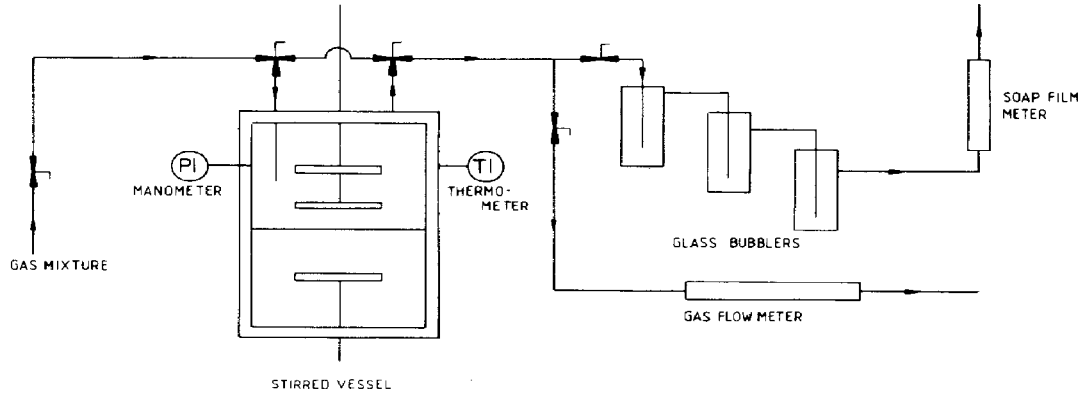

Fig. 1. Experimental set-up for the measurements of the gas-phase mass-transfer coefficients.

The influence of the hydrodynamics was studied by varying the stirrer speed, the height of the stirrer above the gas–liquid surface, the stirrer type (turbine and propeller) and the gas flow rate. In our gas-phase experiments, the liquid phase was a 3 M H_2SO_4 solution which was stirred (about 1 rps) independently of the gas phase.

At the start of each experiment the reactor was filled with the sulphuric acid solution. After closing the reactor the gas mixture was fed to the system at the desired pressure and flow rate. The stirrer speed was also set to the desired value. The product gas was divided into two parts: one part was fed to the gas flow meter and the other part to the glass bubblers in order to determine the composition of the product gas. The inlet and the outlet gas flow and gas composition could be determined in this way.

A 50-fold variation in D_g was obtained by using NH_3 and NH_2CH_3 as the solute gases, and H_2, He, Ar, N_2 and C_2H_6 as the inert bulk gases. The pressures ranged from 127 kPa up to 1.01 MPa. The concentration of the solute gas of the product gas at the inlet was always kept below 3.2% vol. to achieve completely gas-phase controlled mass transfer. A soap-film metered bleed of the product gas was scrubbed in three sequentially sintered glass bubblers containing 500 cm^3 0.01 M H_2SO_4 in order to determine the concentration of the solute in the product gas. To check for possible slip of the solute gas, experiments with two glass bubblers were carried out; these experiments gave the same results as those with the three bubblers. Therefore it can be concluded that the concentration of solute in the product gas was detected correctly. NH_3 and NH_2CH_3 were analysed by standard acid–base titration with 0.01 M NaOH.

Complete ideal mixing of the gas phase was assumed and therefore the gas-phase mass-transfer coefficient was calculated from the stationary mass balance for the gas phase:

$$\phi_g^{in} C_g^{in} - \phi_g^{out} C_g^{out} = k_g A C_g^{out}. \quad (9)$$

The physical properties of the systems used were estimated according to the methods given by Prausnitz *et al.* (1977). Viscosities and densities were calculated for the actual gas mixtures according to Prausnitz *et al.* (1977). Since some of the experiments were carried out at elevated pressures a correction for the non-ideality of the gas phase was necessary. For this correction the Redlich–Kwong equation of state was used.

3.3. *Measurement of the liquid-phase mass-transfer coefficient*

The liquid-phase mass-transfer coefficient was determined by physical absorption of CO_2 into water and of N_2O into aqueous alkanolamine solutions. To avoid gas-phase mass-transfer resistance, single component gases of a high purity were used. For the CO_2–H_2O system, changes in liquid-phase diffusivity were obtained by temperature variation. The rate of reaction between CO_2 and H_2O is much too low to enhance the mass-transfer rate for all the temperatures used. Moreover, the conversion of CO_2 was low and therefore neglected in the mass balance (Pinsent *et al.*, 1956). For the N_2O–alkanolamine systems, changes in diffusivity were obtained by variation of concentration and type of amine. The following amines were used: diethanolamine (DEA), methyldiethanolamine (MDEA) and diisopropanolamine (DIPA). Contrary to the experiments for the determination of k_g, the k_l experiments were carried out batchwise with respect to both the liquid and gas phases.

The experiments were carried out in a 97 mm i.d. and 1.30 l double-wall thermostated all-glass stirred vessel equipped with four equally spaced vertical glass baffles (20 mm) (see Fig. 2). Six turbine impellers (2 blades, 67.2 mm diameter, 10 mm height) were placed on one shaft. By changing the liquid level in the reactor the stirrer action ranged from just one stirrer to three stirrers in the liquid. Also experiments with one of the stirrers in the gas–liquid interface were of course possible. A more detailed description of a similar reactor is given by Blauwhoff *et al.* (1984).

As the experiments were carried out batchwise with respect to both phases, only two aspects of the hydrodynamics were considered to be relevant, i.e. the speed and the position of the stirrers. For the calculation of the mass-transfer coefficient, complete mixing

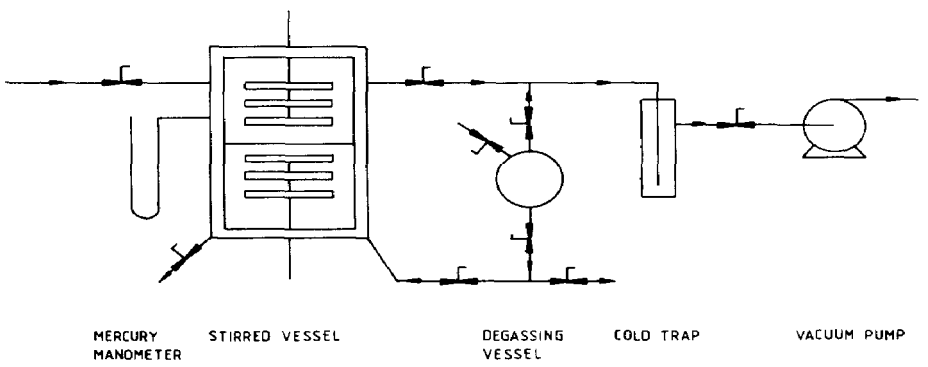

Fig. 2. Experimental set-up for the measurements of the liquid-phase mass-transfer coefficients.

of the liquid bulk was assumed. The unstationary absorption rate is then given by:

$$V_g \frac{dC_g^b}{dt} = -k_l A(He C_g^b - C_l^b) \quad (10)$$

where

$$He = C_l^i / C_g^i. \quad (11)$$

This equation can be solved using the following boundary conditions:

$$C_g^b = C_g^o \quad \text{at } t = 0 \quad (10a)$$

$$V_g C_g^o = V_g C_g^b + V_l C_l^b \quad (10b)$$

$$C_g^\infty = V_g C_g^o / (He\, V_l + V_g). \quad (10c)$$

Integration of eq. (10) gives:

$$\ln\left[\frac{C_g^b - C_g^\infty}{C_g^o - C_g^\infty}\right] = -\frac{(He\, V_l + V_g)(k_l A)t}{V_g V_l}. \quad (12)$$

From eq. (12) the mass-transfer coefficient can be calculated from the decrease in concentration with time, which is constantly monitored by reading the system pressure.

For the analysis and correlation of the experimental data, the physical properties of the systems used are required. These physical properties are given in the Appendix. The absorption of CO_2 into water was studied for temperatures between 18 and 82.5°C; the N_2O experiments were carried out at 25°C. A 15-fold variation in D_l was thus obtained. The maximum stirrer speed at which, according to the eye, a completely smooth interface could be maintained was about 1.2 rps depending on the viscosity of the system.

At the start of each experiment, the liquid was degassed in a separate vessel in order to remove all inert gases. The degassed liquid was introduced under vacuum into the stirred vessel and a vapour–liquid equilibrium was allowed to establish. After equilibration, CO_2 or N_2O was introduced within a few minutes into the vessel until a desired pressure (concentration) level was obtained. The pressure in the vessel was measured with a mercury manometer and read with a cathetometer to ± 0.01 mm Hg. By recording the initial and final pressures (P^0 and P^∞), the solubility of the gas in the liquid was also determined using the following relation:

$$He = \left(\frac{P^0 - P^\infty}{P^\infty}\right)\left(\frac{V_g}{V_l}\right). \quad (13)$$

4. RESULTS

4.1. *Measurements of the gas-phase mass-transfer coefficient*

For the investigation of the hydrodynamic properties of the experimental set-up, NH_3 was absorbed from N_2 into a H_2SO_4 solution and the following parameters were varied: the gas flow rate, the stirrer speed and the total pressure. The position of the stirrer was varied slightly but no influence was found.

In Fig. 3, *Sh* numbers obtained with the propeller stirrer at two stirrer speeds and at a pressure of about

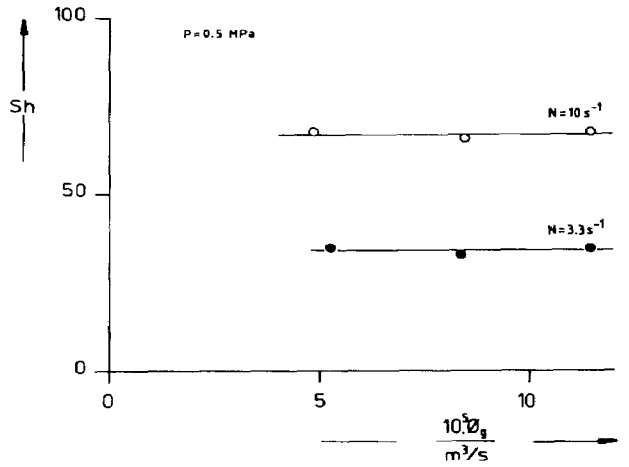

Fig. 3. Effect of the gas flow rate on *Sh* (NH_3–N_2 into H_2SO_4) at 0.5 MPa; the volume flow rate is referred to 0.1 MPa and 298 K.

0.5 MPa are presented for different gas flow rates. The results of these experiments indicate that the values of k_g or *Sh* at constant stirrer speeds are independent of the gas flow rate for the regime studied, which also supports the assumption of ideal mixing . Therefore the main set of experiments was carried out at average gas flow rates of 7.5×10^{-5} m^3/s, corresponding to gas-phase residence times between 40 and 50 s.

The influence of the stirrer speed on k_g was evaluated at three pressures (0.13, 0.5 and 1.0 MPa). In Fig. 4 the results of the experiments with NH_3–N_2 are presented. The relative increase of the mass-transfer coefficient with stirrer speed is equal for all three pressures, i.e. $k_g \propto N^{0.63}$. The concentration of NH_3 in the gas phase at the steady state ranged from 0.27% vol. up to 1.97% vol. At high pressures (i.e. 1 MPa) it is not possible to carry out representative experiments at high stirrer speeds without disturbing the gas–liquid interface. At these high pressures the gas phase interacts severely with the liquid phase, indicating that the pressure increase changes the hydrodynamics of the gas phase although the stirrer speed remains constant. In Figs 5, 6 and 7, photographs are shown of the reactor at a stirrer speed of 20 rps at 0.1 MPa (Fig. 5), 0.5 MPa (Fig. 6) and 1.0 MPa (Fig. 7). These photographs confirm that (at a constant stirrer speed) the hydrodynamic properties of this system change drastically with pressure. In Table 2 the maximum allowable turbine stirrer speeds are summarized. From this table it can be concluded that the critical *Re* number for an undisturbed interface is about 16,000. Although no thorough investigation of this phenomenon was carried out, it is quite clear that the gas-phase hydrodynamics cannot be assumed constant at a fixed stirrer speed or superficial gas velocity when the viscosity or density (pressure) is changed. This is in contradiction with what is stated by Tamir and Merchuk (1978, 1979) and Yadav and Sharma (1979).

The results are correlated with a dimensionless equation, as given in eq. (5). For each system one set of

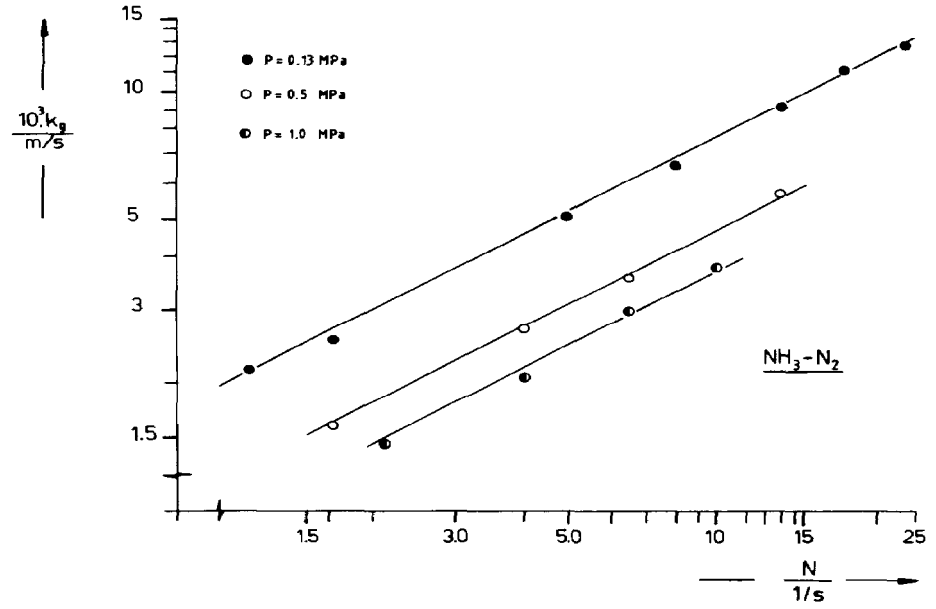

Fig. 4. Effect of the stirrer speed and pressure on k_g (NH_3–N_2 into H_2SO_4).

constants is needed because the *Sc* number does not change in the experimental pressure range. For the gas-phase system NH_3–N_2, the average *Sc* number is 0.69 for all experiments with only very small deviations (< 1.0%). The data shown in Fig. 4 are plotted in Fig. 8 as *Sh* numbers as a function of the corresponding *Re* numbers. The following equation

$$Sh_g = 0.302\, Re_g^{0.63} \qquad (14)$$

correlates all the data for the turbine stirrer within

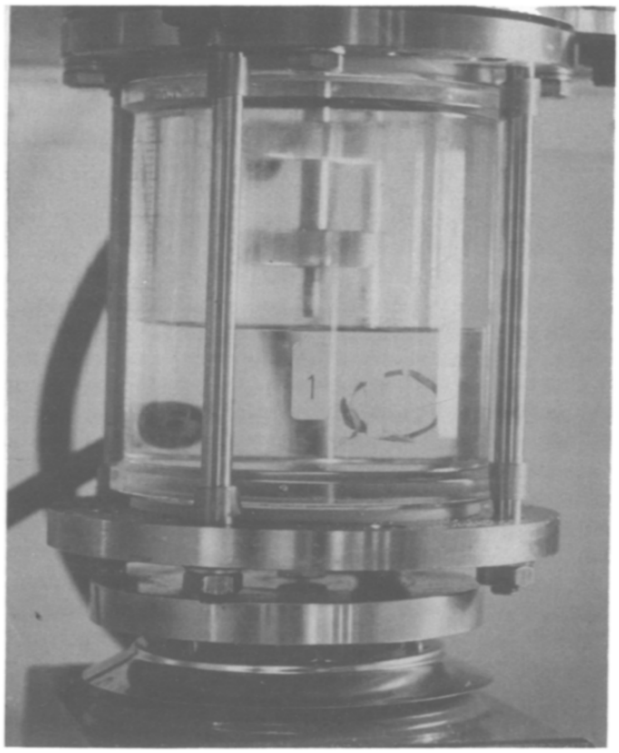

Fig. 5. Gas–liquid interaction in the reactor vessel at *P* = 0.1 MPa and 20 rps.

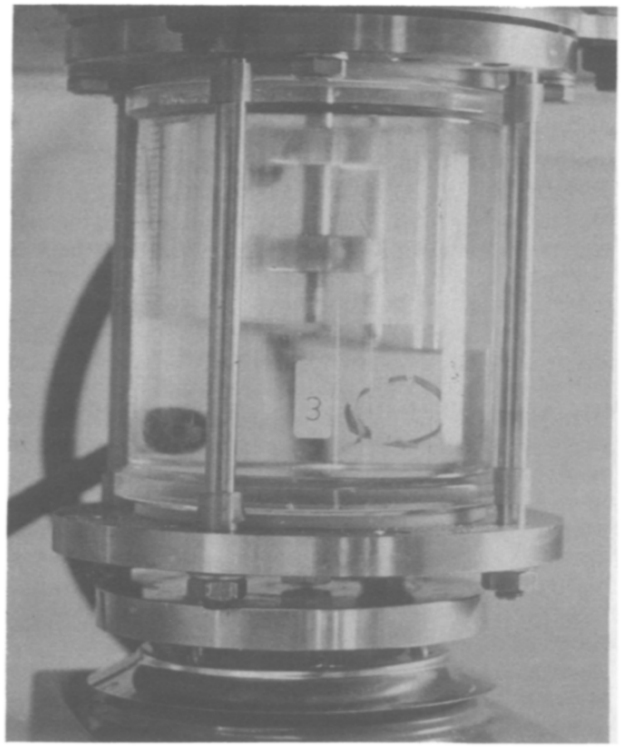

Fig. 6. Gas–liquid interaction in the reactor vessel at *P* = 0.5 MPa and 20 rps.

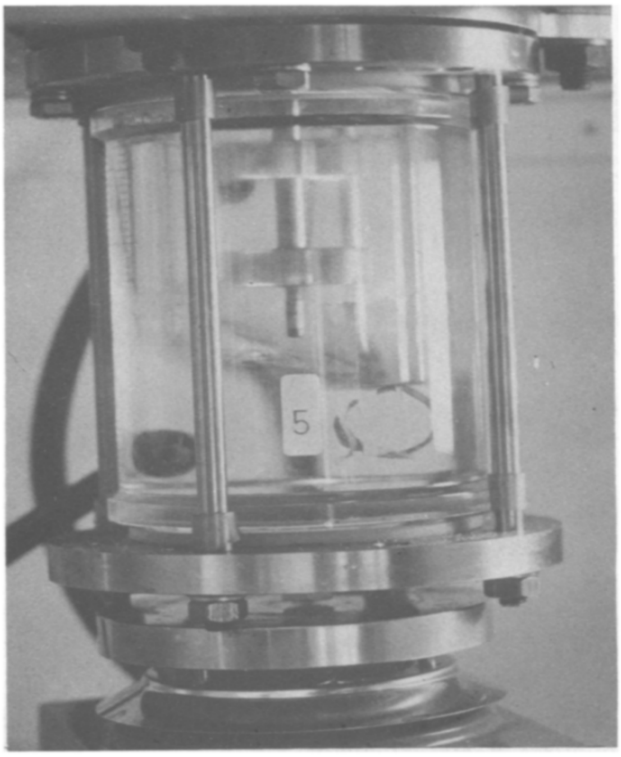

Fig. 7. Gas–liquid interaction in the reactor vessel at P = 1.0 MPa and 20 rps.

5%. In Table 3 the overall results for the other systems are presented. From Table 3 it can be ascertained that the influence of the hydrodynamics, i.e. the Re number, on mass transfer, i.e. the Sh number, for both types of stirrer remains practically constant at the process conditions studied. This means that it is possible to separate the influence of the hydrodynamics (Re number) from the influence of diffusivity on mass transfer.

The exponent $c4$ of the Sc number is determined from all the results according to:

$$Sh/Re^{c3} = c5Sc^{c4} \tag{15}$$

with $c3 = 0.63$ for the turbine stirrer and $c3 = 0.68$ for the propeller stirrer. In Table 4 the average values of Sc and Sh/Re^{c3} are presented. Note that these average values given represent at least four independent measurements. In Fig. 9 the results for both stirrers are presented. The values for $c4$, calculated with a linear regression technique from the data in Table 4, are 0.50 for the turbine stirrer and 0.49 for the propeller stirrer. These values clearly indicate that the dependence of the gas-phase mass-transfer coefficient on diffusivity is proportional to $\sqrt{D}$ for this type stirred vessel. This is in excellent agreement with the Higbie penetration theory or Danckwerts' surface renewal model. Yadav and Sharma (1979) also found that the gas-phase mass-transfer coefficient varies with the square root of diffusivity. As they did not take the changes of the hydrodynamics into account, this result seems to be fortuitous. If we apply the same approach as that of Yadav and Sharma to the present data and the dependence of the gas-phase mass-transfer on diffusivity is evaluated for one stirrer speed, say 10 rps (assuming that the hydrodynamics for all the systems and pressures are constant according to Yadav and Sharma), the gas-phase mass-transfer coefficient would be proportional to $D^{1/3}$. It is not possible to explain this result with the mass-transfer models proposed; however, a good correlation of the available data is obtained with this procedure. For other stirrer speeds an identical conclusion is possible.

A correct way of correlating the experimental data is the use of a dimensionless equation as suggested by Gilliland and Sherwood (1934) and Hikita and Ishikawa (1969). With this approach it is possible to take the influence of the hydrodynamic properties and diffusivity into account separately.

4.2. *Measurements of the liquid-phase mass-transfer coefficient*

Because the stirred vessel has to be filled for each experiment, it is important to know whether the stirrer position relative to the interface has an influence on the liquid-phase mass-transfer coefficient measured. Therefore this aspect was studied for the system CO_2–H_2O at two temperatures, 293 and 303 K, respectively. The effect of the liquid level on k_l was investigated for three relative stirrer positions:

Position 1 : one stirrer completely in the liquid and one stirrer in the gas–liquid interface.

Table 2. Maximum allowable stirrer speeds in the gas phase for the turbine impeller

System	N (s^{-1})	P (MPa)	$(Re_g \text{ number})_{max}$
NH_3–N_2	10	1.0	18,000
NH_3–H_2	No maximum stirrer speed detected	1.0	> 5400
NH_3–Ar	8.33	1.0	16,000
NH_3–C_2H_6	6.67	1.0	26,000
NH_3–He	No maximum stirrer speed detected	1.0	> 5100
NH_2CH_3–N_2	10	1.0	18,000
NH_2CH_3–He	No maximum stirrer speed detected	1.0	> 4200

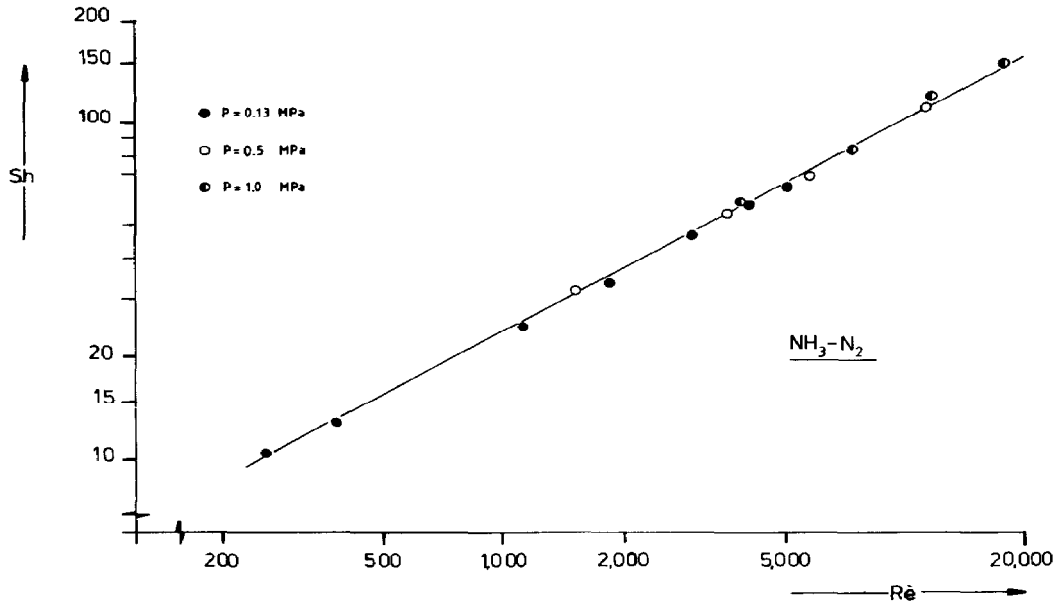

Fig. 8. Sh_g as a function of Re_g for the turbine stirrer.

Table 3. Overall results of the gas-phase mass-transfer experiments

System	Stirrer	$c3$	$Sc_{average}$	Sh range	Re range
NH_3–N_2	Turbine	0.63	0.69	12.4–148	250–18,000
NH_3–H_2	Turbine	0.63	1.34	22.0–104	480–5500
NH_3–Ar	Turbine	0.65	0.64	27.4–140	1250–16,000
NH_3–C_2H_6	Turbine	0.66	0.52	36.0–178	2300–25,000
NH_3–He	Turbine	0.61	1.50	30.1–100	750–5200
NH_2CH_3–N_2	Turbine	0.64	1.07	29.2–221	789–18,000
NH_2CH_3–He	Turbine	0.62	2.05	32.0–111	600–4300
NH_2CH_3–N_2	Propeller	0.72	1.07	29.0–132	1350–11,000
NH_2CH_3–He	Propeller	0.65	2.06	14.3–51.6	380–2200
NH_3–N_2	Propeller	0.66	0.69	32.8–85.9	2700–11,000

Table 4. Average values of Sh/Re^{c3} and Sc numbers

System	Stirrer	P (MPa)	Sh/Re^{c3}	Sc
NH_3–N_2	Turbine	0.13	0.30	0.69
		0.50	0.30	0.69
		1.00	0.31	0.68
NH_3–H_2	Turbine	0.50	0.44	1.33
		1.00	0.45	1.34
NH_3–Ar	Turbine	0.13	0.30	0.65
		0.50	0.31	0.64
		1.00	0.32	0.64
NH_3–C_2H_6	Turbine	0.13	0.28	0.51
		0.50	0.31	0.52
		1.00	0.31	0.53
NH_3–He	Turbine	1.00	0.46	1.48
NH_2CH_3–N_2	Turbine	0.13	0.44	1.08
		0.50	0.44	1.07
		1.00	0.45	1.06
NH_2CH_3–He	Turbine	0.50	0.57	2.06
		1.00	0.58	2.05
NH_2CH_3–N_2	Propeller	0.50	0.23	1.07
NH_2CH_3–He	Propeller	0.50	0.26	2.02
NH_3–N_2	Propeller	0.50	0.15	0.69

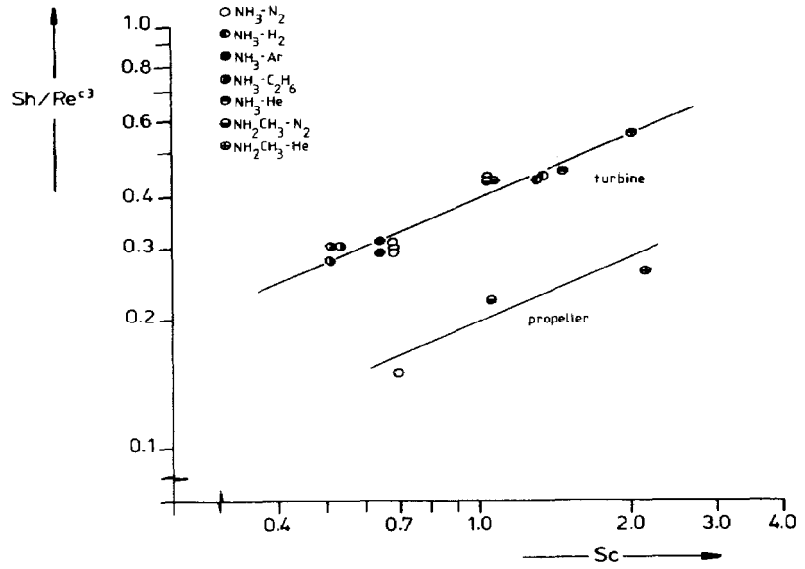

Fig. 9. Overall results of the gas-phase experiments. Sh/Re^{c3} as a function of Sc.

Position 2: two stirrers completely in the liquid.

Position 3: two stirrers completely in the liquid and one stirrer in the gas–liquid interface.

The results are presented in Fig. 10. For 293 K, positions 2 and 3 only and for 303 K all three positions were studied. From this figure it appears that there is no significant difference in the results for the different stirrer positions, so the overall conclusion is that the position of the stirrer has no influence on the liquid-phase mass-transfer coefficient for this particular stirred vessel and in the regime studied. It should be noted that the reactor is operated batchwise with respect to the liquid phase and that in general this conclusion is not applicable for reactors where the liquid phase is operated continuously. In the latter case, there is often a pronounced effect of the stirrer position on k_l.

Next the influence of the stirrer speed on the liquid-phase mass-transfer coefficient was studied. Only four of a total of 30 series are presented in Fig. 11 as typical examples. From the results it can be derived that the

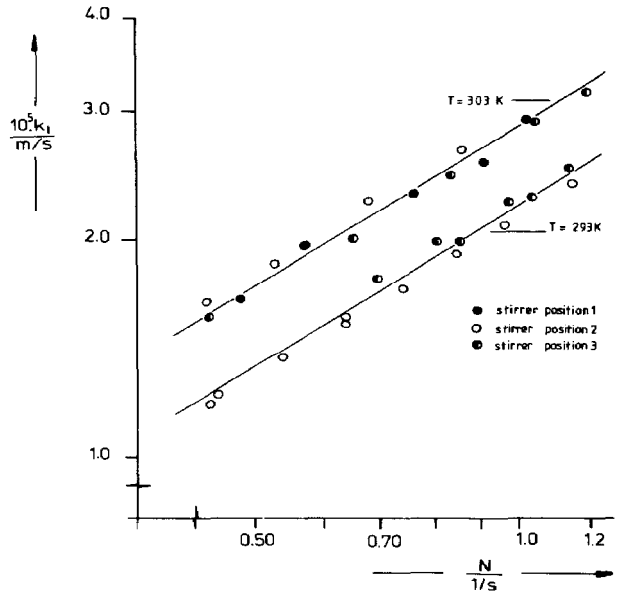

Fig. 10. Experimental results of the influence of the stirrer position.

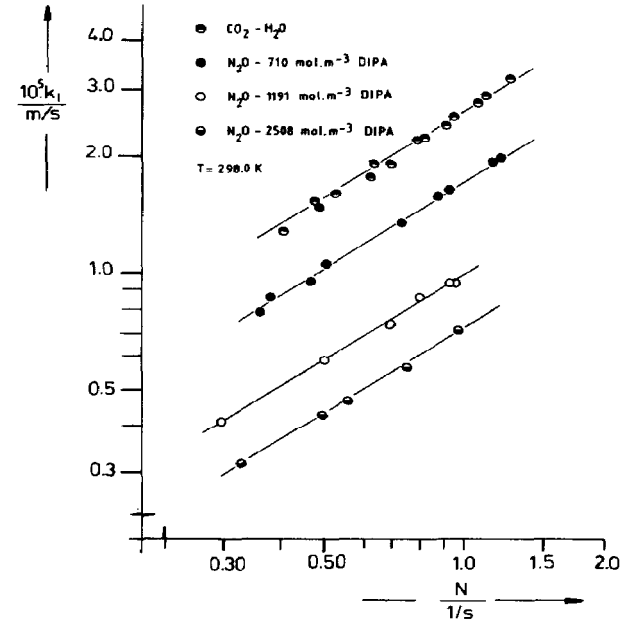

Fig. 11. Experimental results of some typical systems.

dependence of the mass-transfer coefficient on the stirrer speed can be described by the following relation:

$$k_l \propto (N)^{0.72}$$

and that the influence of the stirrer speed is identical for all systems in the experimental range studied. In Table 5 the overall results are presented. Similar to the procedure for the gas-phase experiments, dimensionless numbers are introduced and the influence of diffusivity on the mass-transfer coefficient is described by

$$Sh/Re^{c3} = c2Sc^{c4} \tag{16}$$

with $c3 = 0.72$. All the average values of Sh/Re^{c3} are based on at least four measurements. In Fig. 12 the result of this procedure is presented graphically.

From Fig. 12 it is clear that there is no constant dependence of Sh/Re^{c3} on diffusivity, i.e. Sc number. For high Sc numbers, the dependence tends to a value proportional to $\sqrt{D}$, which may be regarded as a penetration model mass-transfer behaviour. For low Sc numbers, the dependence is about proportional to $Sc^{1/3}$ or $D^{2/3}$ and this seems to be an asymptotic value. Overall it can be concluded that there is a transition in the dependence of the mass-transfer coefficient on diffusivity from proportional to $\sqrt{D}$ (for high Sc numbers) to $D^{2/3}$ (for low Sc numbers) for this particular stirred vessel in the experimental range studied.

4.3. *Interpretation of the liquid-phase mass-transfer experiments according to the multi-parameter models*

As pointed out before, from the mass-transfer models proposed, only the multi-parameter film-penetration models (Dobbins, 1956; Toor and Marchello, 1958) and the interfacial turbulence models (King, 1966; Harriott, 1962; Kishinevskii and Pamfilev, 1949; Kishinevskii, 1955; Kishinevskii and Serebrianskii, 1956) are able to explain the change in dependence of the mass-transfer coefficients on diffusivity. The film-penetration model describes the mass transfer with a gradual change from the penetration to the film model with dependences proportional from $\sqrt{D}$ up to a limit dependence to D. This phenomenon has not been observed for the experiments mentioned above as an asymptotic dependence to $D^{2/3}$ is found and therefore this model is not capable of describing the present results satisfactorily.

The interfacial turbulence model of King (1966)—a combination of unsteady diffusion and eddy diffusion—describes the mass transfer into a surface element by the following equation:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial y}\left[(D + ay^n)\frac{\partial C}{\partial y}\right] \tag{17}$$

Table 5. Overall results of the liquid-phase mass-transfer experiments

System	T (K)	[Amine] (mol/m^3)	$c3$	Sc number	$Sh/Re^{0.72}$	Range
CO_2–H_2O	291	—	0.71	653	1.97	
CO_2–H_2O	292	—	0.72	621	2.01	
CO_2–H_2O	293	—	0.70	592	1.96	
CO_2–H_2O	298	—	0.73	465	1.85	
CO_2–H_2O	303	—	0.71	372	1.75	
CO_2–H_2O	308	—	0.73	299	1.62	$392 \leqslant Sh \leqslant 1245$
CO_2–H_2O	313	—	0.72	244	1.46	
CO_2–H_2O	318	—	0.70	201	1.39	
CO_2–H_2O	323	—	0.69	166	1.28	$1550 \leqslant Re \leqslant 12{,}471$
CO_2–H_2O	333	—	0.71	117	1.18	
CO_2–H_2O	344	—	0.76	83.5	1.04	
CO_2–H_2O	356	—	0.73	58.0	0.871	
N_2O–DIPA	298	289	0.75	591	1.91	
N_2O–DIPA	298	710	0.72	915	2.19	
N_2O–DIPA	298	1758	0.73	3820	4.65	
N_2O–DIPA	298	1901	0.71	4585	5.07	$355 \leqslant Sh \leqslant 1171$
N_2O–DIPA	298	2070	0.71	5726	5.50	
N_2O–DIPA	298	2393	0.73	8802	6.40	$179 \leqslant Re \leqslant 4799$
N_2O–DIPA	298	2508	0.71	10054	7.11	
N_2O–DIPA	298	2804	0.73	13487	8.29	
N_2O–DEA	298	1408	0.74	1327	2.84	$569 \leqslant Sh \leqslant 1973$
N_2O–DEA	298	1556	0.69	1463	2.98	
N_2O–DEA	298	2346	0.76	2407	3.34	$540 \leqslant Re \leqslant 4548$
N_2O–MDEA	298	197	0.72	574	2.13	
N_2O–MDEA	298	585	0.73	757	2.23	$413 \leqslant Sh \leqslant 988$
N_2O–MDEA	298	618	0.72	776	2.30	
N_2O–MDEA	298	1244	0.71	1274	2.75	
N_2O–MDEA	298	1679	0.75	1839	3.02	
N_2O–MDEA	298	1871	0.70	2167	3.30	$1091 \leqslant Re \leqslant 4870$
N_2O–MDEA	298	1907	0.66	2234	3.17	

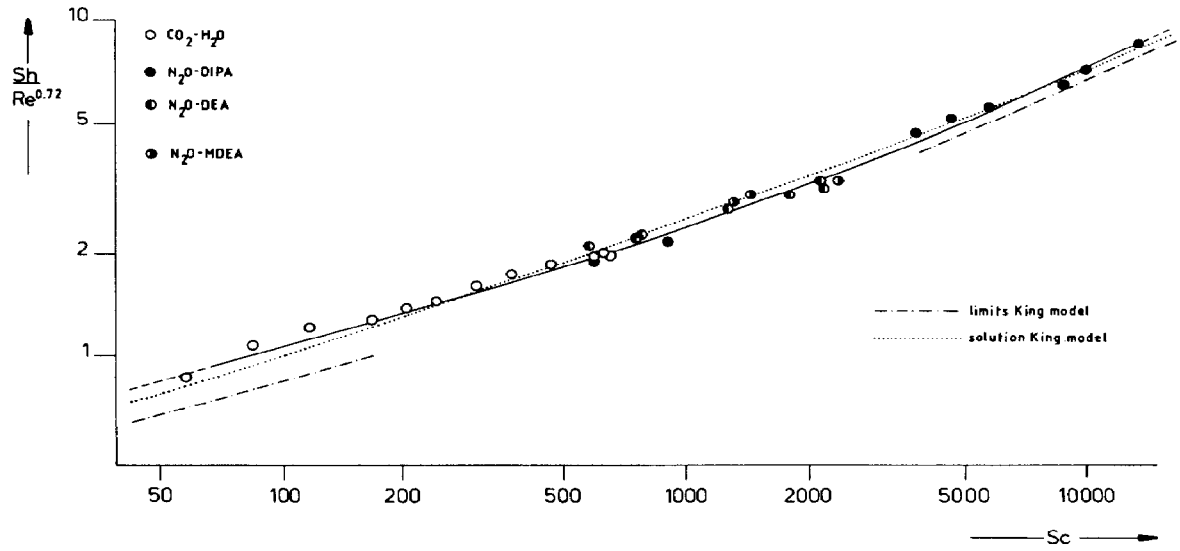

Fig. 12. Overall results of the liquid-phase experiments. Sh/Re^{c3} as a function of Sc.

in which ay^n represents the eddy diffusivity as a function of the distance, y, from the interface. With boundary conditions:

$$C = C_l^b, \quad t = 0 \quad \text{and } y > 0 \tag{17a}$$

$$C = C_l^i, \quad t = 0 \quad \text{and } y = \infty \tag{17b}$$

$$C = C_l^b, \quad t > 0 \quad \text{and } y = 0. \tag{17c}$$

There is no general analytical solution for this differential equation. King (1966) partly solved eq. (17) with dimensional analysis and the introduction of two dimensionless parameters:

(1) the dimensionless mass-transfer coefficient:

$$\Psi = \frac{k_l}{a^{1/n} D^{1-1/n}} \tag{18}$$

and

(2) the dimensionless surface age:

$$\tau = \frac{a^{2/n} t}{D^{(2/n)-1}}. \tag{19}$$

Furthermore, he assumed that $\psi = f(\tau, n)$.

There are two particular limit solutions for eq. (17). The first solution is the case where the surface age is large. In this case, steady-state mass transfer occurs and k_l is independent of the surface age, τ. For this situation Ψ is constant and equal to:

$$\Psi = \left(\frac{n}{\pi}\right) \sin\left(\frac{\pi}{n}\right). \tag{20}$$

This means that k_l is proportional to $D^{1-1/n}$.

The second solution is for relatively low a or t at any n. The term ay^n then becomes negligible compared to the diffusivity D, and mass transfer can be described by the classical penetration theory. In the region between these limit solutions there is a transition where the dependence changes from proportional to $\sqrt{D}$ (low τ) up to $D^{1-1/n}$ (high τ). The King model is therefore able to explain the transition in dependence of the mass-transfer coefficient on diffusivity from the penetration model to a mass-transfer model where the dependence is proportional to $D^{1-1/n}$ (our results indicate that $n = 3$, $k_m \propto D^{2/3}$).

As pointed out above, it is not possible to solve the King model analytically for all values of n, therefore in the transition region a numerical solution of the King model should be used. In Fig. 13 the results from our numerical solution method (Cornelisse *et al.*, 1980) are given for some values of n. The results are presented by the dimensionless mass-transfer coefficient, Ψ, as a function of the dimensionless surface age, τ, according to King (1966). However, time-averaged values are used instead of instantaneous values of k_l and t. Therefore $\tau = (a^{2/n}\theta)/(4D^{(2/n)-1})$, with θ equal to the average age of the surface elements.

In order to compare the King model with the results shown in Fig. 12 values for the parameters a, n and θ are needed. These parameters depend on the hydrodynamics and the physical properties of the system. The value of n can be determined from Fig. 12 using the observation that for low Sc numbers the value of τ is high and consequently one limit solution of the King model is almost reached. The asymptotic dependence of Sh on Sc is 1/3 and therefore the value of n equals 3. An expression for θ can be found for large Sc numbers (low values for τ) if the second asymptotic solution is reached, the penetration model, and if the following correlation is valid:

$$Sh = c5 Re^{0.72} Sc^{0.50}.$$

A similar procedure is applied for a for small Sc numbers when the other limit solution of the King model is reached (Ψ = constant) using the correlation:

$$Sh = c6 Re^{0.72} Sc^{0.33}.$$

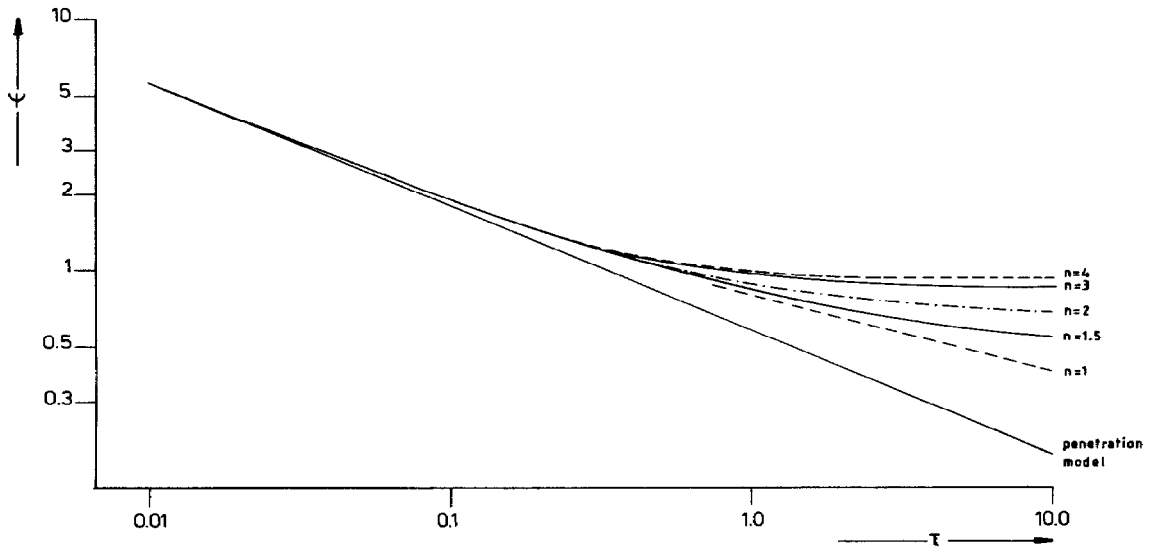

Fig. 13. Numerical solution of the King model for some values of n.

The values of $c5$ and $c6$ are obtained by means of an optimization technique (maximum likelihood principle) (Klaus, 1981). The results are presented in Fig. 14. The fitted values of $c5 = 0.064$ and $c6 = 0.181$, if the experimental data are analysed according to the King model. From Fig. 14 it can be concluded that the King model is able to correlate the dependence of the mass transfer on diffusivity over the whole range of conditions, as measured in our stirred vessel. However, for Sc numbers lower than the values presented here, the dependence of the mass transfer on diffusivity may still increase further and then a higher value of n (e.g. $n = 4$) might even better fit the results. Unfortunately lower Sc numbers are extremely difficult to obtain with our technique. In Fig. 12 the limits and the solution according to the King model are also presented.

Davies *et al.* (1964), Kozinsky and King (1966) and Linek *et al.* (1970) found that the dependence on diffusivity was proportional to $\sqrt{D}$ at a constant stirrer speed (constant hydrodynamic conditions). In terms of the King model this indicates that the values of τ must be small. The only variation in the system studied was the solute gas; therefore, according to eq. (19), the value of τ is constant in view of the hydrodynamical properties as characterized by a and t, and the only change in τ is brought about by a change in the diffusion coefficient. The value of τ does not change for $n = 2$ and in the case of $n = 3$, τ changes by a factor of 1.5 for a variation in diffusivity of a factor of 3.4 which is the maximum range in Kozinsky and King (1966). Therefore it is difficult to determine accurately the value of n of the King model, especially for low values of τ where the King model is close to the penetration limit for all values of n. So it is very well possible that with their experimental set-up small values of τ occur and that the dependence of the mass-transfer coefficient is

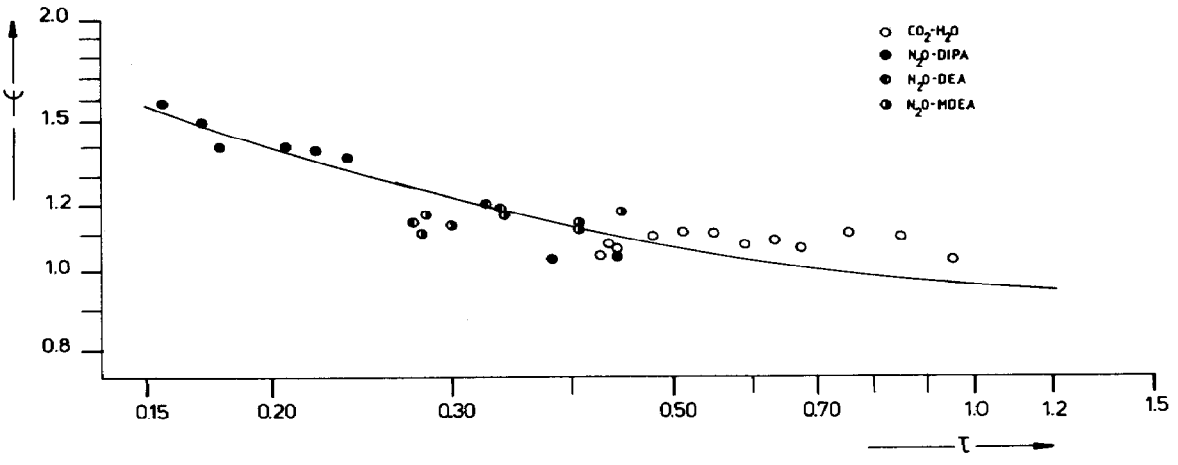

Fig. 14. Comparison between the King model and the experimental data.

proportional to the square root of diffusivity although the actual value of n approaches 3. The same conclusion holds for the work of Tavares da Silva and Danckwerts (1968).

Hikita and Ishikawa (1969) and Machon and Linek (1974) found that the dependence of the mass-transfer coefficient on diffusivity was proportional to $D^{0.67}$. This indicates that $n = 3$ and that their values of τ are high. As stated before, the value of τ is dependent on a, t (brought about by changes in, for instance, the viscosity, density or stirrer speed, geometrical factors) and D, and although the experimental set-up looks similar, τ could be relatively high (Hikita and Ishikawa, 1969; Machon and Linek, 1974) or low (Davies *et al.*, 1964; Kozinsky and King, 1966; Linek *et al.*, 1970) depending on the experimental conditions and geometrical factors. In the present work, however, an important variation in τ was obtained (see Fig. 14) and therefore the value of n can be roughly estimated. The King model is able to explain all the seeming discrepancies which appear from the literature data and also explains the change in dependence on diffusivity for the new data presented here.

Once a correlation, as given in Fig. 12, is established for a particular stirred vessel it can be used to measure the diffusion coefficients of gases into liquids. For example, it is possible to determine accurately the diffusion coefficients of N_2O in aqueous alkanolamine solutions in the stirred vessel studied from simple absorption experiments. These diffusion coefficients may be used for the determination of the diffusion coefficient of CO_2 in the same alkanolamine solutions via the CO_2–N_2O analogy (Laddha *et al.*, 1981). This type of data is required in our study of the coabsorption of CO_2 in alkanolamines while aiming at the selective removal of H_2S.

The advantages of this technique over the classical laminar jet or laminar film are as follows:

(1) less process liquid is needed;
(2) shorter experimental times are possible;
(3) the values of the solubility, He and diffusivity can be determined separately in one single experiment; and
(4) it is easier to handle at high temperatures and pressures.

There are two slight disadvantages: (1) the reactor should be calibrated over a wide range of Sc numbers; and (2) unexpected changes in hydrodynamical behaviour (Marangoni effects, change in dependence on rps) may effect the outcome of the experiments. However, this latter point must be checked for all systems involving gas–liquid mass transfer.

5. CONCLUSIONS

On the basis of the present study, the following conclusions can be drawn for gas–liquid mass transfer in a stirred vessel with a horizontal, smooth interface:

(1) For a correct study of the influence of diffusivity on the mass-transfer coefficient, the dependence of the mass-transfer coefficient on the hydrodynamics and physical properties should be investigated separately.
(2) The hydrodynamics of a system will change when the physical properties, e.g. density of the gas phase, are altered even if constant stirrer speeds are maintained. This has to be taken into account for proper evaluation of mass-transfer experiments.
(3) For the gas phase, the mass transfer in a stirred vessel can be described by the penetration or surface renewal model over a wide range of conditions. The dependence of the mass-transfer coefficient is proportional to $\sqrt{D}$.
(4) Mass transfer in the gas phase at elevated pressures can be correlated with the same dimensionless equation as that given for atmospheric conditions, taking into account the changes of Re.
(5) For the liquid phase, the mass-transfer dependence on diffusivity in stirred vessels is more complex. Therefore the mass transfer should be described by a multi-parameter model. The King model is capable of explaining the observed transition in the dependence of the mass-transfer coefficients (or Sh numbers) on diffusivity and can also account qualitatively for the apparent discrepancies in the literature data. From the present study and the literature review it appears that mass transfer in the liquid phase is proportional to D^n, in which n ranges from 1/2 to 2/3.

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NOTATION

A	interfacial area, m^2
a	constant in eddy diffusivity expression, see eq. (17)
C	concentration, mol/m^3
$c1, \ldots, c6$	constants
D	molecular diffusivity, m^2/s
d_s	stirrer diameter, m
He	dimensionless solubility
k	mass-transfer coefficient, m/s
N	rotation speed, s^{-1}
n	exponent on distance in eddy diffusivity expression
P	pressure, Pa
r	exponent of D
R	gas constant ($= 8.3143$ J/mol/K), J/mol/K
Re	Reynolds number, defined by eq. (7)
S	specific contact area, m^{-1}
s	random surface renewal rate, s^{-1}
Sc	Schmidt number, defined by eq. (8)
Sh	Sherwood number, defined by eq. (6)
T	temperature, K

t	time, s
V	volume, m^3
y	distance normal to interface, m

Greek letters

ϕ	flow rate at standard pressure and pressure, m^3/s
δ	film thickness, m
θ	uniform surface age, s
ρ	density, kg/m^3
η	viscosity, kg/m s
Ψ	dimensionless mass-transfer coefficient, defined by eq. (18)
τ	dimensionless surface age, defined by eq. (19)
[ ]	concentration, mol/m^3

Subscripts

i	interface
b	bulk
g	gas phase
l	liquid phase
m	mass transfer
o	at time $t = 0$
∞	at time $t = \infty$
in	incoming
out	outcoming

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APPENDIX

Physico-chemical properties of the system CO_2–H_2O

Most of the physical properties of CO_2 and H_2O which are necessary for the evaluation of the experiments are taken from literature sources. For the solubility our own measurements are added. The following properties were required:

(a) Liquid density—taken from *Handbook of Chemistry and Physics* (1977).

(b) Liquid viscosity—taken from *Handbook of Chemistry and Physics* (1977).

(c) Diffusivity of CO_2 in H_2O—this is summarized in the following equation:

$$D_{CO_2} = 2.35 \times 10^{-6} \exp(-2119/T/\mathrm{K}) \quad \mathrm{m^2\,s^{-1}} \ (\pm 2\%) \tag{A1}$$

on the basis of the data of literature sources (Thomas and Adams, 1965; Duda and Vrentas, 1968; Unver and Himmelblau, 1964; Scriven, 1956; Clarke, 1964; Davidson and Cullen, 1957; Tammann and Jessen, 1929; Nijsing *et al.*, 1959).

(d) The solubility of CO_2 in H_2O—this is correlated with the following equation:

$$He_{CO_2} = c' RT \exp(2069/T/\mathrm{K}) \quad (\pm 2\%) \tag{A2}$$

in which $c' = 3.30 \times 10^{-5}$ mol/J. The data were compiled from our own measurements and the results published in *International Critical Tables* (1929) and Landolt-Bornstein (1962).

Physico-chemical properties of the system N_2O*-alkanolamines*

The physical properties of the amine solutions were determined by experiments carried out in our laboratory or were published in the literature. All the properties were obtained at 298 K.

(a) The liquid density was calculated with the following equations (Blauwhoff, 1982):

$$\text{MDEA:} \quad \rho = 996.9 + 1.050 \times 10^{-2}[\text{MDEA}] \quad \mathrm{kg/m^3} \tag{A3}$$

$$\text{DEA:} \quad \rho = 997.1 + 1.189 \times 10^{-2}[\text{DEA}] \quad \mathrm{kg/m^3} \tag{A4}$$

$$\text{DIPA:} \quad \rho = 997.5 + 8.37 \times 10^{-3}[\text{DIPA}] \quad \mathrm{kg/m^3}. \tag{A5}$$

(b) The viscosity data for all three systems are presented in Table 6 and the viscosity of the experimental solutions used was determined graphically using these data. Data were published by Blauwhoff *et al.* (1984) and Sada *et al.* (1977, 1978).

Table 6. Viscosity of aqueous alkanolamine solutions at 298 K

System	[Amine] (mol/m³)	$\eta \times 10^3$ (Pa/m)	Reference
MDEA	0	0.8904	*Handbook of Chemistry and Physics* (1977)
	908	1.319	
	1381	1.634	
	1846	2.042	
	2284	2.515	
	2663	3.087	
	3339	4.580	
DEA	498	1.095	Sada *et al.* (1977)
	903	1.236	
	981	1.264	Sada *et al.* (1977)
	1335	1.464	
	1731	1.733	
	1922	1.870	Sada *et al.* (1977)
	2202	2.122	
	2576	2.430	
	2808	2.805	Sada *et al.* (1977)
	3148	3.148	
	3457	3.838	Sada *et al.* (1977)
DIPA	255	0.980	Sada *et al.* (1977)
	486	1.142	Sada *et al.* (1977)
	915	1.544	Sada *et al.* (1977)
	1279	1.957	
	1356	2.080	Sada *et al.* (1977)
	1584	2.418	Sada *et al.* (1977)
	1799	2.828	
	1950	3.098	Sada *et al.* (1977)
	1979	3.180	
	2356	4.131	
	2379	4.244	Sada *et al.* (1977)
	2528	4.771	Sada *et al.* (1977)
	2619	5.136	Sada *et al.* (1977)
	2672	5.377	
	2918	6.603	Sada *et al.* (1977)

(c) The solubility was measured simultaneously with the mass-transfer coefficient. The results are presented in Table 7 together with the data of Sada *et al.* (1977, 1978).

(d) The diffusivity was determined from absorption measurements in a laminar film reactor. The results are presented in Table 8 together with the data of Sada *et al.* (1977, 1978). The term $He\sqrt{D}$ of the experimental solutions was also determined graphically.

Table 7. Solubility of N_2O in aqueous alkanolamine solutions at 298 K

System	[Amine] (mol/m³)	Solubility C_l/C_g	Reference
MDEA	0	0.590	Sada *et al.* (1977)
	614	0.585	
	1236	0.574	
	1253	0.555	
	1640	0.545	
	1665	0.547	
	1731	0.553	
	1871	0.539	
	2422	0.510	
	2517	0.500	

Table 7. (*Contd.*)

System	[Amine] (mol/m^3)	Solubility C_l/C_g	Reference
DEA	449	0.595	Sada *et al.* (1977)
	1397	0.564	
	1418	0.559	
	1556	0.573	
	2026	0.565	Sada *et al.* (1977)
	2290	0.542	
	2313	0.556	Sada *et al.* (1977)
	2360	0.531	Sada *et al.* (1977)
	2389	0.553	
	3081	0.535	Sada *et al.* (1977)
DIPA	225	0.585	Sada *et al.* (1978)
	286	0.607	
	292	0.597	
	486	0.579	Sada *et al.* (1978)
	696	0.565	
	724	0.569	
	915	0.558	Sada *et al.* (1978)
	1356	0.539	Sada *et al.* (1978)
	1584	0.518	Sada *et al.* (1978)
	1758	0.510	
	1901	0.497	
	1950	0.490	Sada *et al.* (1978)
	2070	0.487	
	2379	0.450	Sada *et al.* (1978)
	2393	0.470	
	2508	0.464	
	2528	0.444	Sada *et al.* (1978)
	2781	0.442	
	2826	0.435	

Table 8. $(He\sqrt{D})_{N_2O}$ as a function of the amine concentration and amine type as 298 K

System	[Amine] (mol/m^3)	$(He\sqrt{D}) \geqslant 10^5$ (m/s$^{1/2}$)	Reference
MDEA	0	2.53	Sada *et al.* (1977)
	0	2.55	
	237	2.55	
	407	2.13	
	706	2.17	
	966	2.12	
	1056	2.11	
	1060	2.05	
	1177	1.95	
	1440	1.78	
	1591	1.78	
	1925	1.64	
	2056	1.65	
	2196	1.48	
DEA	540	2.29	
	935	2.14	
	1910	1.76	
	2480	1.61	
DIPA	396	2.33	Sada *et al.* (1978)
	580	2.15	
	830	2.13	Sada *et al.* (1978)
	1107	1.91	Sada *et al.* (1978)
	1280	1.67	
	1410	1.60	Sada *et al.* (1978)
	1663	1.45	Sada *et al.* (1978)
	1753	1.31	
	2330	1.03	
	2763	0.92	